

Dermally Adhered Soil: 1. Amount and Particle-Size Distribution

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ABSTRACT

The risk associated with the dermal absorption of chemicals from contaminated soil is, in part, a function of particle size distribution, as determined by either dry or wet sieving techniques. For the soils tested, the adhered soil fractions were shown to be independent of organic matter content and soil origin. Soil moisture content becomes a factor only for very moist soils. Results show that the adhered fractions of dry or moderately moist soils with wide distributions of particle sizes generally consist of particles of diameters $<63\ \mu\text{m}$. Consequently, dermal absorption experiments using larger size fractions may be of limited relevance to actual situations of soil exposure.

Keywords: Soil Particle-size distributions Wet sieve Adherence Skin

INTRODUCTION

Soil contaminants are potentially bioavailable to humans through particle ingestion, particle inhalation, or dermal contact. Estimating the potential health risk from dermal contact with contaminated soil requires an estimate of the amount of soil (i.e., the soil loading reported in mass per area) that will stick to the skin long enough for the contaminant to transfer from the soil and into the skin (USEPA 2001). Preferential adherence of finer particles has been reported (Duggan et al. 1985; Driver et al. 1989; Sheppard and Evenden 1994; Holmes et al. 1996; Kissel et al. 1996a). Despite this, several dermal absorption experiments using chemicals added to soil (Wester et al. 1990; 1993, 1996; Wester, Maibach, Sedik, Melendres, DiZio, et al. 1992; Wester, Maibach, Sedik, Melendres, Liao, et al. 1992) used soil particle sizes of 48 to 80 mesh (300–180 μm). This size fraction, defined by soil scientists as a medium to coarse sand (Gee and Bauder 1986), is much larger than adhering particles. Because larger soil particles can be significantly different from the smaller particles in terms of composition, especially organic carbon content (Evans et al. 1990; Konen et al. 2003), and thereby contaminant concentration, both the rate and the magnitude of contaminant transfer to the skin could depend on particle size. It is important, therefore, to know the particle-size distribution of adhering soil so that dermal absorption studies can be conducted using relevant soil samples.

Soil adherence on skin has been studied in the laboratory using various procedures to transfer soil to the skin (Que Hee et al. 1985; Driver et al. 1989; Sheppard and Evenden 1994; Holmes et al. 1996; Kissel et al. 1996a) or in the field following a variety of activities (Lepow et al. 1975; Kissel et al. 1996b, 1998; Holmes et al. 1999). Laboratory measurements allow for comparisons of soil properties, whereas field studies better represent actual exposure situations. Table 1 lists a summary of results from some of these studies. Driver et al. (1989) measured adherence by weighing soil samples before and after a volunteer agitated his hand in the sample

for 30 s. The soils were either unsieved or dry sieved into $<250\text{-}\mu\text{m}$ or $<150\text{-}\mu\text{m}$ fractions. Kissel et al. (1996a) and Holmes et al. (1996) measured adherence to palms using a hand-press protocol. In these studies, soil loading per unit area was calculated by estimating the surface area of the total hand area although loading was primarily on the palm side only (Kissel et al. 1996a). The average soil loading was between about 0.3 and 1.4 $\text{mg}\cdot\text{cm}^{-2}$ for the fraction less than 150 μm ; it increased with moisture content and decreased with increasing particle size.

Kissel et al. (1996a) also measured the particle-size fractions (<65 , 65–135, 135–250, 250–425, and $>425\ \mu\text{m}$) of the adhering soil, which was collected using a washing procedure and then dried before dry sieving. In soils with low moisture content (less than 6% moisture) collected from 3 different sites (classified as sand, loamy-sand, and silt-loam soils), they observed that the mass adherence was predominantly attributable to the fraction less than 135 μm or even less than 65 μm . They also observed that the adherence of larger particles increased when the moisture content of these same 3 soils was increased (14–19% moisture). Consistent with these results, in their study of 11 different soils, Sheppard and Evenden (1994) reported that dry particles less than about 50 μm were enriched in the adhering soil relative to the whole soil. Que Hee et al. (1985) observed no relationship between particle size and skin adherence in their investigations of house dust dry-sieved into 6 fractions (<44 , 44–149, 149–177, 177–246, 246–392, and 392–833 μm). However, Que Hee et al. measured skin adherence only once for each size fraction and therefore could not statistically test for differences. They reported a loading of $1.55 \pm 0.83\ \text{mg}\cdot\text{cm}^{-2}$ (mean \pm standard deviation) for all sieve fractions.

Because soil particles can disaggregate in water, it is impossible to know the size of the original particles that adhere to the skin from the particle-size distribution measured by wet sieving alone. Kissel et al. (1996a) examined this question by comparing the dry-sieve fractions of the adhering soil to the bulk soil treated following the same procedure (i.e., dispersed in wash water and then dried). At low moisture content, they found that the adhering soil was

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Table 1. Summary of results from prior studies of soil adherence on skin

Study source	Driver et al. (1989)	Kissel et al. (1996) ^a	Holmes et al. (1996) ^a
Soils	13 samples from 5 sites	5 sites	3 sites
Description ^b	PF and WS	PF	WS (<2 mm)
Moisture content	Air dried not measured	12–18%	<2%
Adherence ^c in mg·cm ⁻²	1.40 ± 0.45 for <150 µm 0.98 ± 0.27 for <250 µm 0.58 ± 0.24 for whole soil	0.66 (0.34–1.30) for <150 µm 2.96 (0.32–8.31) for 150–250 µm 5.98 (0.39–10.2) for >250	0.29 sand 0.37 loamy sand 0.59 silt loam
	0.62 (0.42–0.76) for <150 µm 0.34 (0.24–0.49) for 150–250 µm 0.14 (0.06–0.34) for >250	0.31 (0.22–0.54)	1.63 (0.39–3.09)
	<2%	0.1–9%	13–15%
			1.6 sand 3.7 loamy sand 0.25 silt loam

^a Adherence values were based on total hand area, although loading was primarily on the palm side only. For comparisons with Driver et al. (1989) and the current study, the adherence value should be doubled.

^b PF = prefractionated soil samples; WS = whole soil.

^c Mean ± standard deviation for all soils or mean (range from low to high).

enriched in small particles compared to the bulk soil for the sand and loamy-sand soils. For the silt-loam soil, the smallest particle-size fraction was present at about the same amounts in the adhering and bulk soils. For soils at high moisture content, the larger particle sizes were enriched in the adhering soil compared with the bulk soil. While these results clearly demonstrated that the particle sizes of the adhering soil often differ from that of the bulk soil, distinguishing whether the cause was preferential adherence of certain particle sizes or disaggregation of adhering larger particles is not possible.

The research reported here is an investigation of the soil characteristics that affect particle adherence to human skin, especially particle-size distribution of the original soil, organic carbon content, and soil moisture. The experimental design allowed investigation of the major sources of variability in dermal soil adherence, including individual subject and day-to-day variability. Adhesive tape was used to remove adhered soil so that the soil mass could be measured. The effects of particle-size distribution and organic carbon content were studied by washing using deionized water, similar to other studies (Holmes et al. 1996; Kissel et al. 1996a). In contrast to the study by Kissel et al. (1996a), this investigation compared the adhering particle-size distribution measured by wet sieving, with dry and wet sieving of the original soil sample, to account for both the amount and the size fraction that disaggregated during the washing process. The effect of soil moisture content on adherence was examined to a limited extent.

EXPERIMENTAL METHODS

Soil sampling

The 2 soil samples used in this research were collected from the upper 25 cm of the soil profiles after removal of the sod layer. The Colorado State University (CSU) soil, a clay loam (classified using percent sand vs percent clay according to Gee and Bauder [1986]), was collected from the CSU Agricultural Research Station in Fort Collins, Colorado, USA. The Iowa State University (ISU) soil is a silty-clay loam collected from the ISU Field Extension Experimental Laboratory in Ames, Iowa, USA. The soils were dry sieved (using a 0.6 × 0.6-m suspended shaker with 6 × 6-mm screen openings) to break up large clods and remove rocks. Samples passing through the sieve were then coned and quartered, and one-quarter was hand sieved using a brass 2 mm (No. 10) sieve. The >2-mm fraction of the ISU soil, which formed small balls because the soil was moist, was lightly broken apart using a mortar and pestle and then resieved. This process was repeated until all the soil had been sieved. Both soils were stored in large plastic containers covered with a porous plastic mesh that allowed them to air dry. The <2-mm soil fraction (the original or bulk soil) was subsampled either by coning and quartering or by using a Humboldt splitter (model H-3980; Humboldt Manufacturing, Norridge, IL, USA).

Percent moisture

Soil moisture was varied by placing soil samples in closed containers with a relative humidity of 100% (high), 45% (medium), or 15% (low), obtained using solutions of distilled water, saturated KNO₃, and saturated LiCl, respectively (Lide 1984). The laboratory temperature was kept at 19 to 21 °C. The percent moisture of the soil (i.e., the mass of water per mass of dry soil) was determined in triplicate by placing a

measured mass of soil in a 100 °C oven and drying to a constant mass. For the 2 soils studied here, moisture contents of soil samples equilibrated at low, medium, and high humidity were 1% to 2%, 3% to 4%, and 9% to 10%, respectively. In some experiments with the CSU soil, moisture of the air-dried soil was measured after the initial processing without any adjustment, and the sample was stored in a tight container to maintain constant moisture. The moisture content of this soil (4.70%) was about half as high as for the high-humidity samples and slightly higher than for the medium-humidity samples.

Bulk soil particle-size analysis

A dry-sieve particle-size analysis was conducted just prior to each adherence experiment by placing a 2- to 5-g subsample of soil in 7.6-cm-diameter stainless-steel sieves (ASTM E-11 specification) that were stacked as follows: 500 μm (No. 35), 250 μm (No. 60), 125 μm (No. 120), 63 μm (No. 230), 38 μm (No. 400), and 25 μm (No. 500). The top cover and bottom pan were also stainless steel. The stack was shaken, using a RX-29 ROTAP, for 15 min. The resulting size fractions were then placed in preweighed plastic Petri dishes, dried in a 50 °C oven, and then weighed to determine the mass of each fraction. Additional samples of the bulk soil were also taken for percent moisture determination.

At the start of each particle-size experiment, a soil suspension for a wet-sieve analysis of the bulk soil was prepared by placing a 1.5- to 5-g subsample of the bulk soil into a beaker and adding approximately 100 mL of distilled water. Following the completion of the adherence experiments, this soil suspension was poured through the previously described sieve stack. Each sieve was rinsed with distilled water to ensure passage of particles smaller than the sieve size. Once the smaller particles had been transferred to the next sieve, the retained fraction was rinsed into a preweighed Petri dish and dried in a 50 °C oven to constant weight. The <25- μm fraction was centrifuged at 3,000 g for 30 min using a Marathon 12KBR Fisher Scientific centrifuge. The samples were decanted, dried in a 50 °C oven, and then weighed. The aqueous suspensions from the multiple-subject adherence experiments were treated in a similar manner.

Dermal adherence of soils

Healthy adult human volunteers (108 total; ~one-third female; mostly students between 18 and 30 y of age; all but 10 were Caucasian) participated in this study (approved by the University of California, San Francisco, CA, USA, Institutional Review Board) after written consent was obtained. Unless noted otherwise, volunteers participated in the study without any preparatory skin treatment, such as hand washing. Experiments with individual subjects were conducted to ascertain the magnitude of and variations in adhered soil mass per unit area of skin. Wash samples from a group of volunteer subjects (either 20 or 30) were combined to obtain a large enough sample to determine the particle-size distributions and organic carbon contents of adhered soil compared with the bulk soil. For both sets of experiments, small, open-ended containers of known cross-sectional area (average was 13.6 cm^2) were partially filled with bulk soil, and the open end was covered by the subject's palm. The container and palm were inverted 10 times, allowing the soil to contact the skin. The container was then removed, and with the inverted palm facing down, the back of the hand was

given a gentle tap to remove loose soil. The remaining soil was removed from the palm with either preweighed commercially available adhesive tape (clear packaging tape such as Scotch® Book Tape 845, 3M, or similar) (for individual subject experiments) or distilled water (for particle-size experiments). The tape-strip method was used only with the CSU soil, whereas the washing method was used with both soils.

For the individual subject adherence experiments, the tape, with the adhered soil, was weighed, and the mass of soil per area was calculated for each sample. Blanks were performed by applying preweighed tape strips to the palms of unwashed hands, without added soil, then removing and reweighing the tape. The value for the blank was determined from the change in mass.

For the experiments using pooled samples from multiple subjects, adhered soil from each subject was washed with deionized water into a single container using a plastic spray bottle. The wash solutions for all subjects, collected over 2 to 5 h, were combined and wet sieved using the technique described previously. During intervals between subjects, soils were stored in the appropriate humidity chamber. An average contact area of the containers was used to determine mass adhered per area.

Carbon analysis

Samples of bulk soil and size fractions of both bulk and adhered soil were ground to homogenize them for total and inorganic carbon analysis, using a CO₂ Coulometer (Coulometrics, Model 5011). Organic carbon content was calculated from the difference between total and inorganic carbon. The performance of the instrument was evaluated by analyzing blanks and standards prepared from known masses of calcium carbonate (CaCO₃).

The total carbon analysis consisted of heating a weighed soil sample in a platinum boat at 900 °C, converting all the carbon in the sample to carbon dioxide (CO₂). The CO₂ was then swept with CO₂-free oxygen into the CO₂ coulometer for quantification. Blanks were performed on an empty boat using the previously described procedure.

For the inorganic carbon analysis, 2 mL of 2 N perchloric acid (HClO₄) were added to a weighed soil sample in a glass tube, converting all the inorganic carbon in the sample to carbon dioxide (CO₂), within 5 min. The CO₂ was similarly quantified using the coulometer. Blanks were run using the previously described procedure on an empty glass tube.

RESULTS AND DISCUSSION

Tape-strip experiments

Table 2 lists the results of the individual tape-strip experiments for the CSU soil, collected from 6 subjects on 5 different days. An average of approximately 0.70 $\text{mg}\cdot\text{cm}^{-2}$ of bulk CSU soil adhered to the subjects' palms, and there was no significant difference between the left and right hand. It can also be seen from Table 2 that there were similar but large average variabilities in the mass per area of adhered soil, both between subjects on a given day ($\pm 52\%$) and for an individual subject on different days ($\pm 50\%$).

To further investigate the amount of variability due to the subjects' hands, a larger number of subjects were tested on a single day in an additional experiment. The results from these 1-d tape-strip experiments are shown in Table 3. A somewhat higher adhered mass per area, 1.14 $\text{mg}\cdot\text{cm}^{-2}$, was obtained for

Table 2. Adherence of Colorado State University (Fort Collins, CO, USA) soil for each subject and day measured by tape stripping^a

	Right hand			Left hand		Both hands	
Day	<i>n</i> ^b	mg·cm ⁻² ^c	<i>n</i>	mg·cm ⁻²	<i>n</i>	mg·cm ⁻²	
1	6	0.806 ± 37%	6	0.789 ± 53%	12	0.797 ± 43%	
2	6	0.578 ± 55%	6	0.695 ± 71%	12	0.637 ± 63%	
3	6	0.611 ± 59%	6	0.628 ± 61%	12	0.620 ± 57%	
4	6	0.636 ± 71%	6	0.876 ± 37%	12	0.756 ± 52%	
5	6	0.686 ± 60%	6	0.709 ± 26%	12	0.697 ± 44%	
				Average coefficient of variation = 52%			
Subject	Days	mg·cm ⁻²	Days	mg·cm ⁻²	Days	mg·cm ⁻²	
1	5	0.694 ± 44%	5	0.976 ± 25%	10	0.835 ± 41%	
2	5	0.594 ± 47%	5	0.659 ± 58%	10	0.627 ± 42%	
3	5	1.07 ± 21%	5	1.02 ± 25%	10	1.04 ± 24%	
4	5	0.546 ± 62%	5	0.518 ± 30%	10	0.532 ± 70%	
5	5	0.574 ± 73%	5	0.449 ± 71%	10	0.511 ± 89%	
6	5	0.501 ± 69%	5	0.818 ± 53%	10	0.695 ± 34%	
				Average coefficient of variation = 50%			
All subjects/all days	30	0.663 ± 54%	30	0.739 ± 49%	60	0.701 ± 51%	

hands that were not washed at the beginning of the experiment. The coefficient of variation for this larger number of subjects, 57%, was similar to the experiments with fewer participants. This suggests that the variability between individuals on a given day, arising from the differences in the subject's skin characteristics, is similar to the variability for an individual subject from 1 day to another.

Table 3 lists the results for an experiment where the subjects washed their hands with soap and water and then rinsed and towel dried them before the soil was contacted. The average mass per area (0.5 mg·cm⁻²) for washed hands is similar to the previous results shown in Table 2 but is considerably less than for the nonwashed hands in this experiment. The lower adhered mass is probably due to the removal of oils from the skin that aid in the adherence of soil particles. Table 3 also reports the results from the blank measurements from unwashed hands, which were at least an

order of magnitude smaller than the mass of adhered soil. Since the focus of this research was to determine the effect of soil characteristics (particle-size distribution and carbon content) on soil adherence to skin under common situations, the subjects' hands were not washed in subsequent experiments.

Particle-size experiments

For the particle-size experiments, the average adherence was determined for a group of subjects (either 20 or 30 individuals) in 3 or 4 repeated experiments (Table 4). Comparing results from the particle-size experiments of the medium hydration (3.81% moisture) CSU soil to the 1-d tape-strip experiments without hand washing (also on the CSU soil with a similar moisture content of 4.70%; Table 3), soil adherence for the particle-size experiments (0.62 mg·cm⁻²) was less than for the 1-d tape-strip experiment

Table 3. Adherence of Colorado State University (Fort Collins, CO, USA) soil measured by tape stripping on multiple subjects in 1 d with and without previous hand washing^a

Study type	Right hand		Left hand		Both hands	
	<i>n</i> ^b	mg·cm ⁻² ^c	<i>n</i>	mg·cm ⁻²	<i>n</i>	mg·cm ⁻²
No washing	36	1.18 ± 50%	36	1.10 ± 63%	72	1.14 ± 57%
Washing	38	0.517 ± 54%	38	0.501 ± 55%	76	0.509 ± 55%
Blank					8	0.0403 ± 109%

^a Soil moisture content was 4.70%.

^b Number of subjects.

^c Mean ± coefficient of variation in %.

Table 4. Soil adherence measured in groups of subjects as a function of 3 hydration levels in 2 soils^a

Soil	Soil hydration					
	Low			Medium		
	% Moisture ^b	mg·cm ⁻² ^b	n ^c	% Moisture	mg·cm ⁻²	n
CSU	1.85 ± 1.6%	0.64 ± 9.4%	80	3.81 ± 10%	0.62 ± 4.8%	100
ISU	1.58 ± 1.1%	0.69 ± 14%	110	3.35 ± 13%	0.67 ± 19%	70
	High					
	% Moisture	mg·cm ⁻²	n			
CSU	9.36 ± 11%	1.47 ± 11%	90			
ISU	10.1 ± 36%	1.36 ± 10%	110			

^a CSU = Colorado State University, Fort Collins, CO, USA; ISU = Iowa State University, Ames, IA, USA.

^b Mean ± coefficient of variation in % for *m* experiments.

^c Either 20 or 30 subjects participated in each experiment; *m* = number of experiments; *n* = total number of subjects who participated in these *m* experiments.

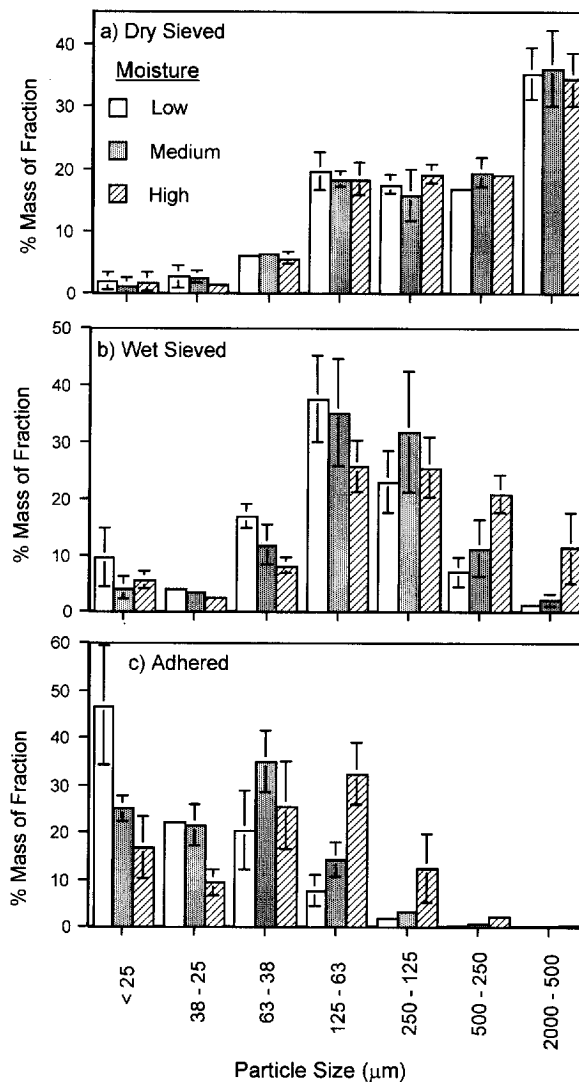


Figure 1. Particle-size distribution of the Colorado State University (CSU, Fort Collins, CO, USA) soil at the 3 moisture contents listed in Table 4 (mean values ± standard deviation).

(1.14 mg·cm⁻²). This difference cannot be accounted for by soil lost during sieving since the percent recovery for bulk wet sieving is greater than 90% (data not shown) for both soils at all moistures. The blank measurements also indicate that particles and oils present on the hands before washing cannot explain the difference. A possible explanation is that tape stripping might be more efficient at removing soil than is washing. As will be discussed in a subsequent section, the particle-size distribution for the adhered soils is quite different compared to the bulk soil. Most important, the adhered soil contains only a small mass of larger particles. Therefore, small losses of a few large particles, each of which contributes a large amount of mass, could also explain the difference between the mass per area determined for the tape-stripping and washing procedures. This might also explain the large amount of variability that is apparent in the results listed in Tables 2 and 3. Certainly, the results listed in Tables 1 and 2 are consistent in both the amount and the variation of adherence with those observed in previous studies (Table 1).

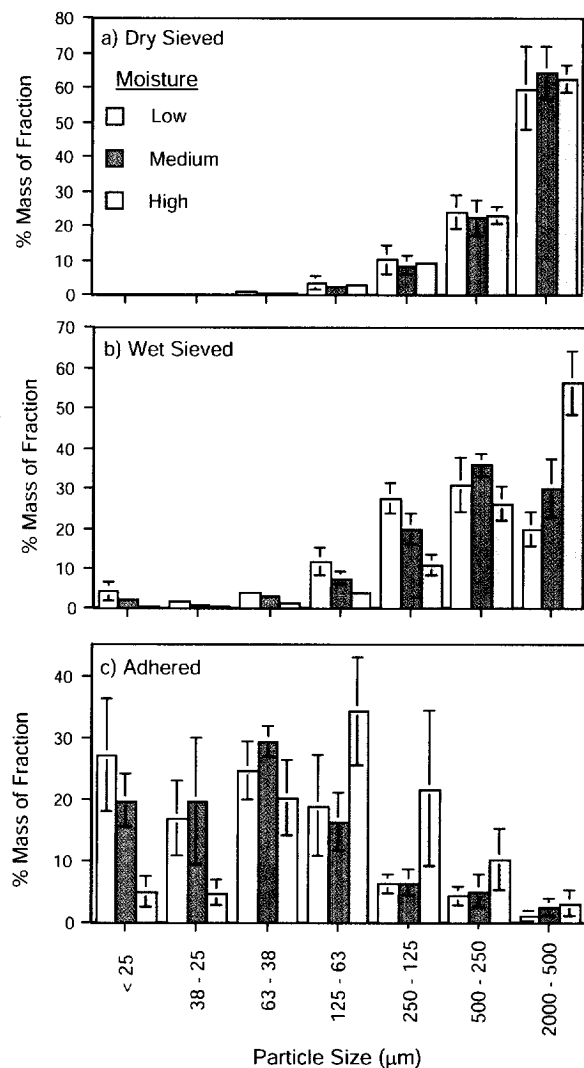


Figure 2. Particle-size distribution of the Iowa State University (ISU; Ames, IA, USA) soil at the 3 moisture contents listed in Table 4 (mean values \pm standard deviation).

As shown in Table 4, the average adhered mass per area is similar for both soils prepared with both low and medium levels of hydration. Further investigation with different soil types, especially with different particle-size distributions, is required to confirm that soil characterization (e.g., organic carbon content, mineralogy, clay characterization) may not be necessary to estimate the mass adhering to the skin, as suggested by these findings. The adhered mass did increase significantly ($p < 0.0025$) by about a factor of 2 for both soils at the highest level of hydration compared to the lower hydration levels. This is consistent with previous observations (Holmes et al. 1996; Kissel et al. 1996a). It should be noted that this study did not investigate soils with free water.

The size distribution of the soil adhering to human palms was compared with the size distribution of the bulk soil measured by dry and wet sieving. The dry-sieve analysis of the bulk soil represents the particle-size distribution of the soil to which subjects were exposed. Since the adhered soil was wet sieved, which might cause particle disaggregation, samples of the bulk soil were wet sieved for comparison. Three initial

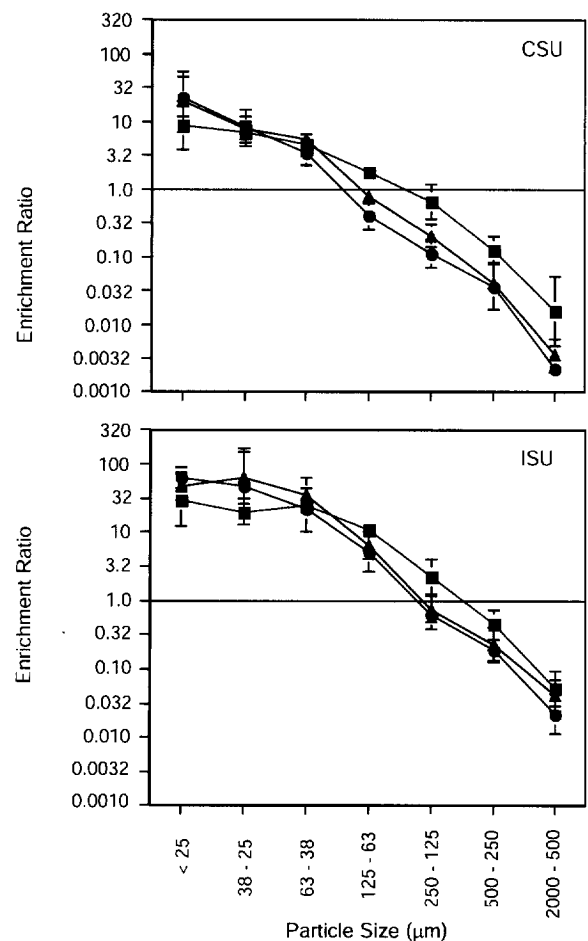


Figure 3. Enrichment of each particle-size fraction in the adhered soil compared with the bulk soil for the Colorado State University (CSU; Fort Collins, CO, USA) and Iowa State University (ISU; Ames, IA, USA) soils at the 3 moisture contents listed in Table 4 (mean values \pm standard deviation): low (\circ), medium (Δ), high (\blacksquare).

percent moistures were compared. These results for 3 hydration levels of the CSU and ISU soils are shown in Figures 1 and 2, respectively. The error bars for the adhered size distributions represent the standard deviation in the number of experiments (m) listed in Table 4. The error bars for the results from dry and wet sieving of the bulk soil were developed from the standard deviations of the determinations for the number of experiments listed in Table 4. Some error bars on these figures are too small to be observed at these scales, which is also true for all the following graphs unless stated otherwise.

For both soils, the bulk soil samples show a shift to smaller sizes with wet sieving. However, the shift is far less significant for the ISU soil than for the CSU soil, suggesting that the particle-size distribution of the ISU soil is more water stable. For both soils, after dry sieving, the percent mass of the largest particle-size fraction (i.e., 500–2,000 μm) was the same for all 3 moisture contents, but after wet sieving, the percent mass of this fraction was highest for the highest-moisture-content soil. However, only the highest-moisture ISU soil was statistically different from the lower-moisture soils. This suggests that the largest particles in the high-moisture-content ISU soil were less prone to disaggregation than in the low- and medium-

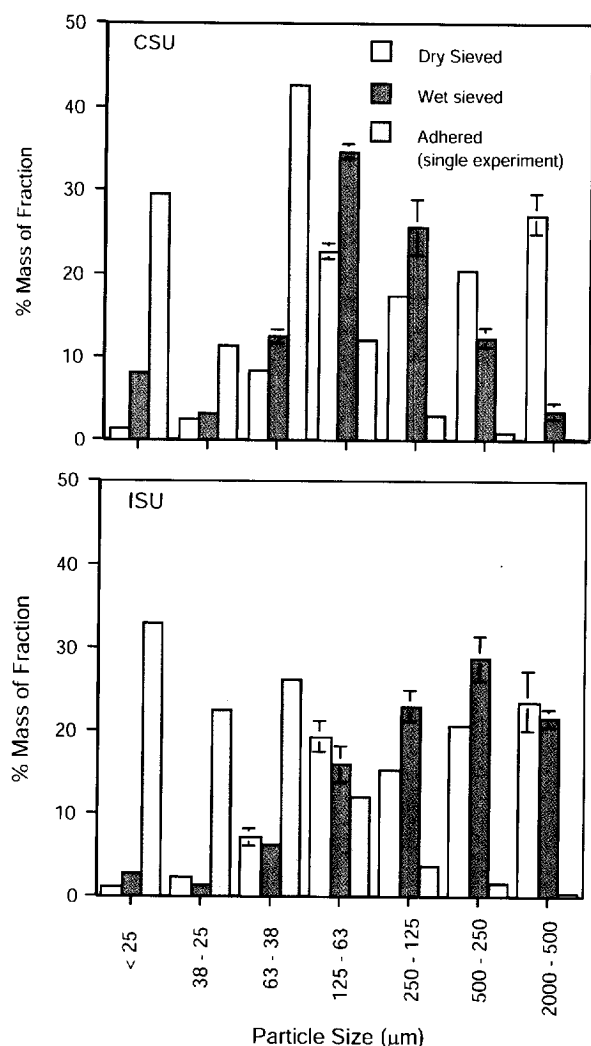


Figure 4. Comparison of the particle-size distributions of the adhered soil to the wet-sieve and dry-sieve bulk soil of the reconstituted Colorado State University (CSU; Fort Collins, CO, USA) and Iowa State University (ISU; Ames, IA, USA) soils at medium moisture content (mean values \pm standard deviation).

moisture soils. It is likely that the normal moisture content for a soil from Iowa is higher than the low- or medium-moisture levels studied. If so, perhaps removal of water from the interior of the aggregates may reduce the hydrogen bonding and thereby decrease resistance to water-mediated disaggregation. An alternative explanation is that the pressure created by capillary uptake of water, which may force the aggregates apart, is greater for the drier soil.

An even greater shift to smaller sizes is observed in the adhered wet-sieve fractions for both soils as compared to the dry-sieve soil. This is consistent with a preferential adherence of the finer soil fractions. A slight increase occurs in the percent mass of the larger particle sizes for the adhered experiments with increasing soil moisture. These results are similar to those reported previously by Kissel et al. (1996a).

Since the soil is adhered dry, washed from the palm, and then wet sieved, it is likely that at least some of the particles in the fine fractions from the adhered soil are due to disaggregation of particles from larger size fractions. This

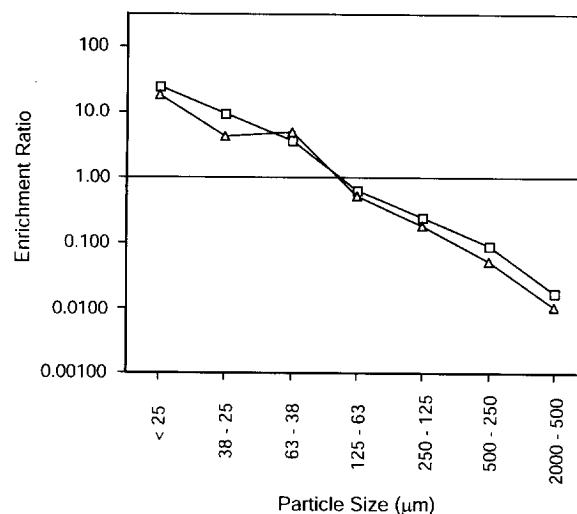


Figure 5. Enrichment of each particle-size fraction in the adhered soil compared with the bulk reconstituted soils at medium moisture content (mean values \pm standard deviation): Colorado State University (CSU; Fort Collins, CO, USA) (Δ) and Iowa State University (ISU; Ames, IA, USA) (\square).

was investigated by wet sieving a series of dry-sieve fractions. The results from this study, which are presented in the companion paper (Choate et al. 2006), indicate that the finer fractions of soil disaggregated less than the larger particles. Furthermore, the adhering particle sizes for these soils are small enough that they display minimal disaggregation in water. Therefore, when the potential for disaggregation was taken into account using algorithms described by Choate et al. (2006), the adhered particle-size distribution did not change significantly.

The enrichment ratio of the adhering particle sizes was determined by dividing the percent mass in the adhering fraction by the percent mass in the corresponding dry-sieve fraction. Figure 3 shows the enrichment ratio plotted as a function of particle-size fraction. All values greater than 1 represent adhered size fractions that were enriched as compared to the amount that was in contact with the skin. As an example, for the CSU soil at medium hydration, the 38- to 63- μm size fraction makes up 35% of the mass of the adhered soil but only 6.5% of the mass of the bulk soil. At this moisture, a higher proportion of this fraction adhered than was represented in the bulk soil.

Reconstituted soil experiments

Enrichment in the adhering particle sizes can be seen in size fractions less than 125 μm for the CSU soil and less than 250 μm for the ISU soil (Figure 3). The cause of this difference between the 2 soils, especially the influence of the differences in the initial dry size distributions of the 2 soils, was also investigated (Choate et al. 2006). This was done by physically combining the dry-sieve fractions of each of the 2 soils in the proportions to match the original CSU soil dry-sieve distribution. In this way, the 2 different soils, now having the same initial bulk dry-sieve size distributions and termed CSU or ISU reconstituted soil, could be compared. The percent moisture for both soils was in the medium range (2.1–4.3%). Size fractionation experiments with the reconstituted bulk soils were performed in triplicate for the dry- and wet-

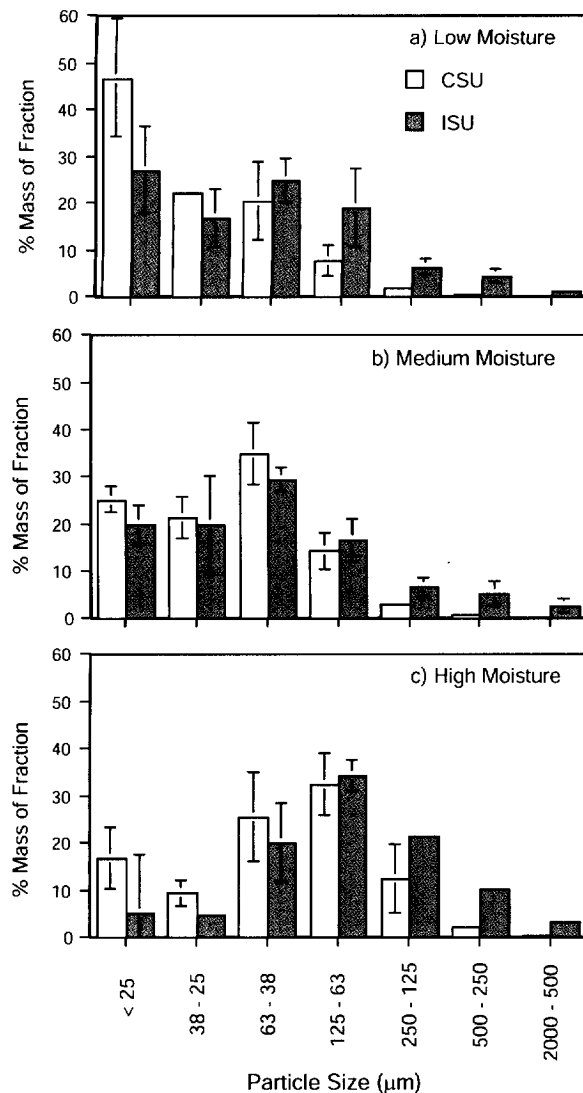


Figure 6. Particle-size distribution in the adhered Colorado State University (CSU) and Iowa State University (ISU) soils at the 3 moisture contents listed in Table 4 (mean values \pm standard deviation).

sieving techniques. However, only 1 multiple-subject adherence experiment with the 2 reconstituted bulk soils was performed. Figure 4 shows that the dry size distributions of the reconstituted bulk soils are statistically the same. Also, the wet distribution of the reconstituted bulk soils follows the same pattern as the original bulk soils (Figures 1b and 2b). The adhered distribution of the reconstituted bulk soils follows the same trend for the CSU soil as the original bulk CSU soil but is shifted to smaller particle sizes compared to the original bulk ISU soil (Figures 1c and 2c). This is not surprising since there is more mass initially in the smaller size fractions of this reconstituted bulk ISU soil compared to the original bulk ISU soil.

Figure 5 shows that when the reconstituted bulk soils have the same dry size distribution, their enrichment in adherence is the same and occurs at less than 63 μm . This leads to the conclusion that particle-size enrichment for these 2 soil types is dependent on the initial soil mass in the dry-sieve fractions and independent of other soil characteristics.

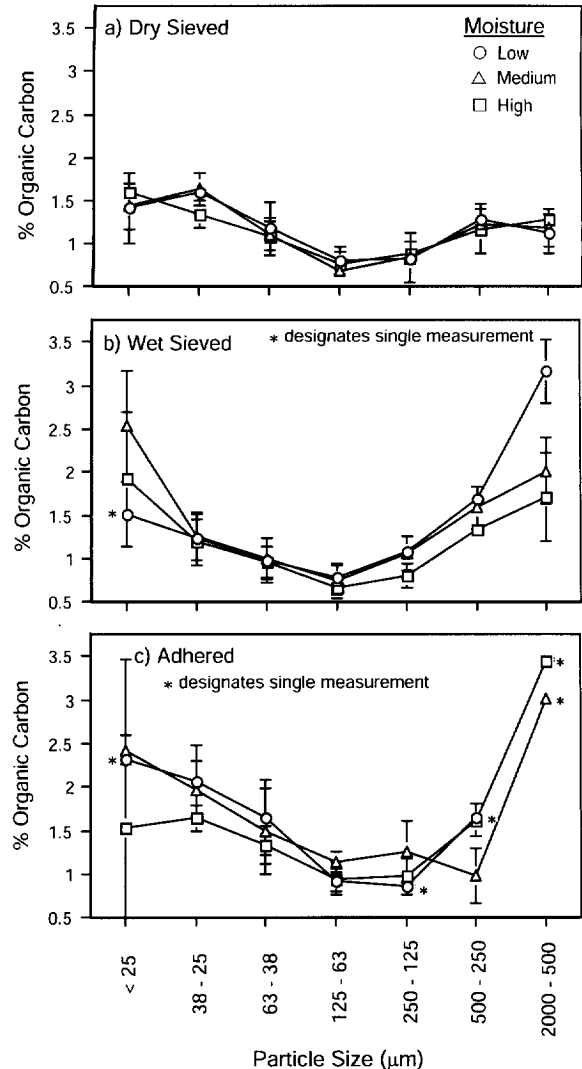


Figure 7. Organic carbon content in each particle-size fraction of the Colorado State University (CSU) soil at the 3 moisture contents listed in Table 4 (mean values \pm standard deviation).

Figure 6, which combines data shown in Figures 1c and 2c, shows that although the particle-size distributions of the original bulk CSU and ISU soils are different, this had little effect on the size distribution of the adhering particles. Although the enrichment factors for the CSU and ISU soil are different, the size distribution of the adhering particles was affected minimally. So, for example, the bulk ISU soil has more large particles than the bulk CSU soil, and, as a result, there was enrichment in particles of 125 to 250 μm for the ISU soil in comparison to the CSU soil. This again confirms that the initial soil size distribution is the single most important factor in influencing enrichment.

Organic carbon

The size distribution and the potential influence of organic carbon on adherence were also investigated. The percent organic carbon distribution for the CSU soil and the ISU soil are presented in Figures 7 and 8, respectively. For both the CSU and the ISU soils, the pattern of organic carbon versus particle size was similar in the wet-sieve bulk soil and the

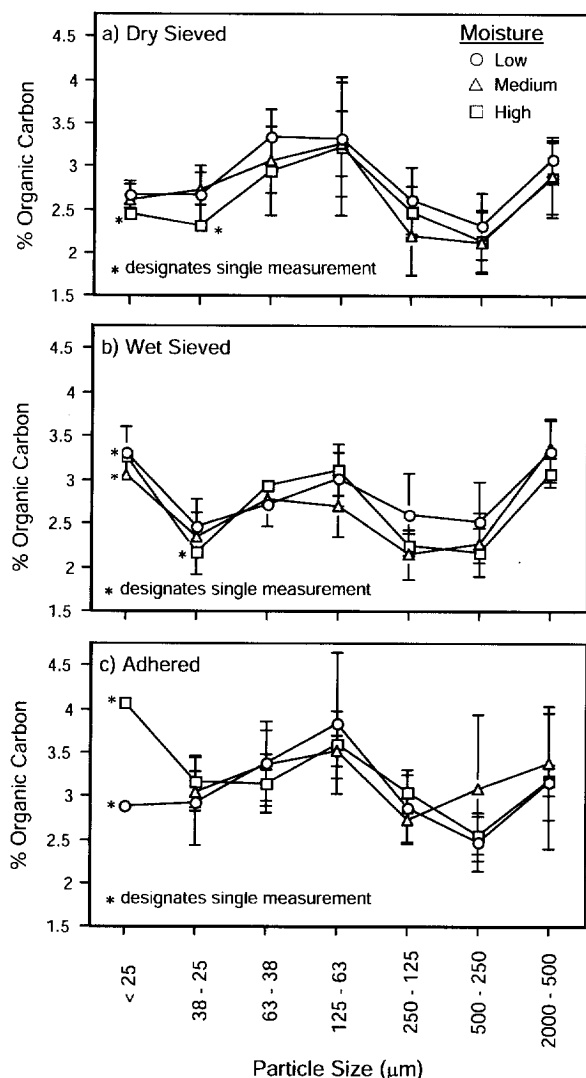


Figure 8. Organic carbon content in each particle-size fraction of the Iowa State University (ISU) soil at the 3 moisture contents listed in Table 4 (mean values \pm standard deviation).

adhered soil. Thus, organic carbon does not enhance the adherence of any of the particle sizes, which is consistent with the results of a study by Holmes et al. (1996).

SUMMARY

The dermal loading of soil ranged from 0.62 to 1.14 mg·cm⁻². It was determined from the tape-strip experiments that the variation between subjects on the same day was similar to the day-to-day variation for each subject.

The size-dependent enrichment of adhered soil particles was identical for the CSU and ISU soils when their dry particle-size distributions were matched. Thus, enrichment was different for these 2 soils because their bulk soil particle-size distributions were different. The enrichment was independent of moisture content for the low- and medium-moisture soils. For the soils with higher moisture content, more of the large particle fractions adhered compared to the low and medium moisture contents. It should be noted that this study did not investigate soils with

free water. Therefore, adherence for very moist soils will require further study.

Organic carbon content, mineralogy, the nature of particle aggregation, and particle-size distribution were different for the CSU and ISU soils (Choate 2002) but had no effect on the particle-size distribution of the adhered soil. It is reasonable to assume that dermal exposure to other soils with wide particle-size ranges similar to these soils will result in similar adhered amounts and particle-size distributions. However, this conclusion may not be appropriate for soils with limited particle-size distributions (e.g., play sand that consists of mostly large particles and an extremely small fraction of small particles). In this case, the large particles may block the small particles, and, consequently, the amount of small particles adhering would be less, making the total amount adhering less. This will require further investigation.

Although soil characteristics such as organic carbon may not be important for adherence, they might be important for dermal absorption of soil-associated contaminants, but there is no information on whether soil particle size affects dermal absorption. Given that the adhering soil is mostly composed of particles <125 μm, the data from Wester et al. (1990, 1993, 1996; Wester, Maibach, Sedik, Melendres, DiZio, et al. 1992; Wester, Maibach, Sedik, Melendres, Liao, et al. 1992), which was from a medium to coarse sand (300–180 μm), may not be representative of dermal absorption in real exposures. The effect of particle size on dermal absorption needs to be studied further.

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REFERENCES

- Choate LM. 2002. The effect of variations in soil organic matter with particle size on organic contaminant sorption and its relationship to dermal exposure [PhD thesis]. Golden (CO): Colorado School of Mines.
- Choate LM, Ranville JF, Bunge AL, Macalady DL. 2006. Dermal adhered soil: 2. Reconstruction of dry-sieve particle-size distributions from wet-sieve data. *Integr Environ Assess Manag* 2:385–390.
- Driver JH, Konz JJ, Whitmyre GK. 1989. Soil adherence to human skin. *Bull Environ Contam Toxicol* 43:814–820.
- Duggan MJ, Inskip MJ, Rundle SA, Moorcraft JS. 1985. Lead in playground dust and on the hands of children. *Sci Tot Environ* 44:65–79.
- Evans KM, Gill RA, Robotham PWJ. 1990. The PAH and organic content of sediment particle size fractions. *Water Air Soil Pollut* 51:13–31.
- Gee GW, Bauder JW. 1986. Particle-size analysis. In: Klute A, editor. *Methods of soil analysis: Part I—Physical and mineralogical methods*. Madison (WI): Soil Science Society of America. p 383–411.
- Holmes KK, Kissel JC, Richter KY. 1996. Investigation of the influence of oil on soil adherence to skin. *J Soil Contam* 5:301–308.
- Holmes KK Jr, Shirai JH, Richter KY, Kissel JC. 1999. Field measurement of dermal soil loadings in occupational and recreational activities. *Environ Res* 80:148–157.
- Kissel JC, Richter KY, Fenske RA. 1996a. Factors affecting soil adherence to skin in hand-press trials. *Bull Environ Contam Toxicol* 56:722–728.
- Kissel JC, Richter KY, Fenske RA. 1996b. Field measurement of dermal soil loading attributable to various activities: Implications for exposure assessment. *Risk Anal* 16:115–125.

- Kissel JC, Shirai JH, Richter KY, Fenske RA. 1998. Investigation of dermal contact with soil in controlled trials. *J Soil Contam* 7:737–752.
- Konen ME, Burras CL, Sandor JA. 2003. Organic carbon, texture, and quantitative color measurement relationships for cultivated soils in north central Iowa. *Soil Sci Soc Am J* 67:1823–1830.
- Lepow M, Bruckmas L, Gillette M, Markowitz S, Robino R, Kapish J. 1975. Investigations into sources of lead in the environment of urban children. *Environ Res* 10:415–426.
- Lide DR, editor. 2005. CRC handbook of chemistry and physics. Boca Raton (FL): CRC. 2544 p.
- Que Hee S, Peace B, Clark C, Boyle J, Bornschein R, Hammond P. 1985. Evolution of efficient methods to sample lead sources, such as house dust and hand dust, in the homes of children. *Environ Res* 38:77–95.
- Sheppard SC, Evenden WG. 1994. Contaminant enrichment and properties of soil adhering to skin. *J Environ Qual* 23:604–612.
- [USEPA] US Environmental Protection Agency. 2001. Risk assessment guidance for Superfund, Volume I: Human health evaluation manual (Part E, Supplemental guidance for dermal risk assessment), interim guidance. Washington DC: Office of Emergency and Remedial Response. EPA/540/R/99/005.
- Wester RC, Maibach HI, Bucks DA, Sedik L, Melendres J, Liao C, DiZio S. 1990. Percutaneous absorption of [14C]DDT and [14C]benzo[a]pyrene from soil. *Fundam Appl Toxicol* 15:510–516.
- Wester RC, Maibach HI, Sedik L, Melendres J, DiZio S, Wade M. 1992. In vitro percutaneous absorption of cadmium from water and soil into human skin. *Fundam Appl Toxicol* 19:1–5.
- Wester RC, Maibach HI, Sedik L, Melendres J, Liao CL, DiZio S. 1992. Percutaneous absorption of [14C]chlordane from soil. *J Toxicol Environ Health* 35:269–277.
- Wester RC, Maibach HI, Sedik L, Melendres J, Wade M, DiZio S. 1993. Percutaneous absorption of pentachlorophenol from soil. *Fundam Appl Toxicol* 20:68–71.
- Wester RC, Melendres J, Logan F, Hui X, Maibach HI, Wade M, Huang K-C. 1996. Percutaneous absorption of 2,4-dichlorophenoxyacetic acid from soil with respect to soil load and skin contact time: In vivo absorption in rhesus monkey and in vitro absorption in human skin. *J Toxicol Environ Health* 47:335–344.